

A cost-effective approach to realization of the efficient methane chemical-looping combustion by using coal fly ash as a support for oxygen carrier

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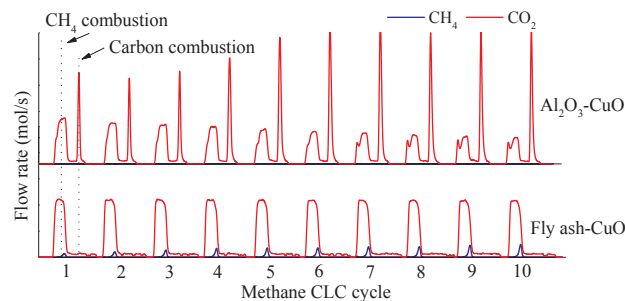
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HIGHLIGHTS

- FA is better than the Al_2O_3 as a support of OC in anti-carbon deposition and durability.
- Iron oxide contained in FA support could provide active oxygen for fuel combustion.
- Partially irreversible CuO decomposition is the main reason for the deactivation of OC.
- The distribution of Cu on the support became more uniform after reaction.

GRAPHICAL ABSTRACT



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ABSTRACT

Developing highly reactive, durable and low-cost oxygen carrier (OC) is the key to the advancement of the chemical looping combustion (CLC) technology that is regarded as the most energy-efficient way for the capture of CO_2 emitted from fossil fuel combustion. In this study, the byproduct from coal power plant, coal fly ash (FA) was utilized as the support for Cu-based OC in methane CLC. Its performance was investigated with a thermogravimetric analyzer and a lab-scale fixed bed reactor. The results showed the FA is better as a support of OC than the most commonly used Al_2O_3 in anti-carbon deposition and thus beneficial to the importance of the stability of FA supported OCs. Among all the tested OCs, the Cu-based OC synthesized with FA support and impregnation method demonstrated the best performance, with CH_4 conversion of 94–100% and CO_2 selectivity of 91–94% in ten cycles at 800–850 °C, and minor deactivation. The characterization of the fresh and spent OCs revealed that the FA support itself could provide 1.0–2.9 wt% active oxygen for fuel combustion, which is enabled by the iron oxide in FA. In addition, the distribution of Cu on the FA support became more uniform on spent OC than on the fresh one. Also, the partially irreversible CuO decomposition into Cu_2O during cyclic tests was the main reason for the deactivation of OC. Thus, Use of FA as an OC support is conducive to both development of cost-effective CLC and an environmental-friendly utilization of waste FA.

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1. Introduction

Since the industrial revolution, fossil fuels have been used to drive unprecedented economic and agricultural growth, which has led to the release of tremendous CO₂ into the atmosphere. CO₂ is considered to be closed with the increasingly concerned global warming [1]. In order to slow down the pace of global warming, the improvement of the energy utilization efficiency and development of substitute energy are critical, so is the development of CO₂ capture and storage (CCS) technology. Among all the CCS technologies such as the pre-combustion capture, post-combustion capture, oxy-combustion and chemical looping combustion, the last one would be a great choice because it does not bring any enthalpy gains [2] and can avoid the energy-intensive processes otherwise needed for CO₂ capture including CO₂ absorption and desorption as well as air separation, thus place CLC as a top of the CCS technology. However, the CLC still cannot be commercialized due to the difficulty in finding efficient oxygen carriers achieving both the high reactivity with fuels and multicycle durability or stability. Also owing to the need for large amount of OC cycled in the system, the cost of the OC is also critical to the application of CLC technology.

To date, more than 700 materials including active metal oxides or supports have been developed for synthesis of OCs [3]. For active metal oxides, particularly transition metal oxides including those Cu, Fe, Co, Ni, Mn based ones have been extensively studied. CuO is considered to be most suitable one that can transport its oxygen to fuels with high reaction rate and oxygen supply capacity without any thermodynamic restriction for complete fuel conversion to CO₂ and H₂O [4,5]. Besides, Cu is a less expensive and more environmental-friendly material than Co and Ni [3,6].

As an indispensable ingredient in the OC preparation, the support is to provide an inert, firm and porous structure for the active metal oxide component, increasing the diffusion capacity and strength of OC, therefore easing the reaction with fuels, avoiding the agglomeration and deactivation. Some commonly tested supports for OCs are SiO₂, Al₂O₃, TiO₂, ZrO₂, MgO, MgAl₂O₄. Besides, industrial byproducts and natural ores can be the supports of OCs to decrease the synthesis cost, they share the same feature that containing part of the inert materials abovementioned, also the active metal oxide contained could provide oxygen for the fuel combustion. Red mud (bauxite residue) is the byproduct of the alumina production industry and containing Al₂O₃, SiO₂, TiO₂, CaO and Fe₂O₃. It was proved to be a good candidate as the OC or support, with high reactivity, stability and high oxygen carrying capacity in CLC [7,8]. The slags from the iron and manganese production industries were evaluated by Fosdahl [9] as OCs, and it was indicated that the silicates and aluminates contained were able to tolerate the high temperature and ensure the multicycle performance of OC. Some natural ores simultaneously contain active metal oxide (Cu/Fe/Mn oxides) and inert support were also intensively studied [10–13]. Tian [13] studied three copper ores and five iron ores e.g. chrysocolla, cuprite, magnetite and hematite for their application in CLC, the result showed good recycle capability for these ores, however the low active metal contents restricted their combustion and reoxidation performances. Besides, Bentonite (mainly containing SiO₂ and Al₂O₃) [14,15], olivine (mainly containing Mg₂SiO₄ and Fe₂SiO₄) [6,9], cement (mainly containing silicate) [16] are all potential cheap materials in CLC.

To further develop a cost-effective support for the OC synthesis, fly ash as one of the byproducts from coal fire power plants is selected as a support for OC in this study. Fly ash is generated at 1200–1700 °C and quite thermally stable, with spherical shape and porous structure. It contains bulk chemical compounds such as SiO₂, Al₂O₃, MgO, and TiO₂, which are potential OC supports [17–20]; in the meantime, the Fe₂O₃ in FA could function as an active metal oxide to provide the lattice oxygen for the fuel combustion [21], other elements such as Na and K are the promoters for the OC [22]. Thus, theoretically, the fly ash should be a promising material as a support for OC synthesis.

Aisyah [23] firstly proposed the utilization of FA as support in OC synthesis and loaded copper, nickel and iron oxides onto the FA and evaluated them in syngas CLC. The results indicated that among the OCs tested, CuO–FA had higher oxygen transfer efficiency and oxygen carrying capacity than other metal oxide–FA OCs did. Skulimowska [24] also evidenced the good performance of FA as a support for CuO in chemical looping with oxygen uncoupling (CLOU), the synthesized OC did not suffer from sintering/deactivation under CLOU tests, showing high and stable activity in 42 cycles. Bao [25] studied the interaction between iron-based OCs and the coal ashes in CLC, and found the ashes containing the active species (such as Fe₂O₃ or CaSO₄) that functioned as oxygen carriers could extend the reduction capacity; while the chemical combination between the ash species and the carrier was unfavorable for the combustion. Hence, the FA was proved to be a suitable support for the OC in gaseous fuel combustion. Beyond that, with the development of solid fuels CLC, more attention was focused on the interaction between the coal/biomass ash with the OC. Ruble [26] investigated the effect of different coal ashes on the performance of OC for coal CLC, and found only lignite ash had a beneficial effect on the ability of OC to oxidize carbon, but all other ashes were detrimental and reduced the ability of the OC to remove carbon from the mixtures. Wang [27] used iron and copper based OC for CLC of an anthracite, and found that the ash reacted with the minerals in coal ash to form Al₂SiO₅ and free Al₂O₃ possibly lowering the reactivity of OC in the further redox cycles. Saha [28] investigated the interaction between the CuO and the mineral inherent in the coal. It was found that the reactivity and oxygen transport capacity of OC was restricted due to the deactivation effect of minerals in coal.

So far, neither performance of FA supported Cu OC for the methane combustion nor the interaction between the Cu and the FA during the methane CLC cycles has been studied yet. Thus, this paper is trying to exam the applicability of FA in the methane CLC, and develop a new cost-effective OC for the CLC technology. Furthermore, since it is estimated that the annual production of the FA worldwide is around 500 million tons [29], thus use of fly ash as a support for CLC could considerably reduce its adverse impact on the environment.

2. Experimental

2.1. Preparation and characterization of OCs

The coal fly ash tested in this study is from the Laramie River Power Station in Wyoming State, USA, a byproduct from the combustion of the Powder River Basin subbituminous coal. The main components in the as-received fly ash were measured by inductively coupled plasma (ICP) and shown in Table 1.

The pretreatment of the fly ash included calcination, grinding and drying. The as-received fly ash was placed in a muffle furnace and calcined at 950 °C for 5 h, to eliminate the carbon residue contained. The obtained sample was then ground to all pass the 125 μm griddle, followed by drying at 110 °C for 2 h in a vacuum oven to remove the moisture absorbed during the grinding. The comparative material is commercial γ-Al₂O₃ (Alfa Aesar, 99.9%, particle size of 40 μm and surface area of 100 m²/g).

The OCs tested in this research were fly ash and Al₂O₃ (marked as FA and AL, respectively) supported CuO, prepared with two methods, mechanical mixture (MM) and wet impregnation (IM). For each OC, the mass fractions of CuO and support in OCs were both 50 wt%. The OC prepared with mechanical mixture method is labeled as FA/AL MM, its

Table 1
Main components in Power River Basin coal fly ash.

Element	Al	Fe	Ca	Mg	Si
Content (mg/g)	72.69	28.18	173.57	27.33	132.51

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